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13. ABSTRACT (Maximum 200 words)

We have developed microscopic theory for analysis of spin excitations in semiconductor nanocrystals and endohedral fullerenes based on atomistic electronic structure calculations. We have made calculations of shape and size dependence of electronic and excitonic g-factors, examined magnetic properties of Mn:ZnS dilute magnetic semiconductor nanocrystals, made atomistic calculation of coherent spin transfer between nanocrystals linked by organic molecules, calculated structural, magnetic and transport properties of a high spin endohedral metallofullerene, Gd@C82, and calculated hyperfine coupling constants for endohedral nitrogen atoms in fullerenes of variable size. Comparison with experimental data is made, and good agreement is found in all cases, validating the microscopic methods developed here.

14. SUBJECT TERMS
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(Continuation Sheet)

Final Progress Report

Statement of Problem Studied

The aim of this project was to develop microscopic theory for analysis and prediction of spin coherences in semiconductor nanostructures and to make applications to specific instances relevant to using spin degrees of freedom in solid state for quantum information processing. In the course of the work we expanded the scope of the project to address also spin degrees of freedom in endohedral metallofullerenes. We developed microscopic methods for calculation of electronic and excitonic g-factors in nanocrystals, an atomistic approach to calculation of coherent spin transfer between coupled nanocrystals, and spin transport calculations for coupled nanostructures.

Summary of Significant Results

1. In the first year of the program, we developed a time-independent semi-empirical tight-binding method for the calculation of the Lande g-factor of semiconductor (specifically CdSe) nanocrystals. This system was of interest to us due to the wealth of time resolved Faraday rotation (TRFR) experimental data which indicated the presence of multiple g-factors for certain nanocrystal size ranges. This study gave qualitative results indicating the shape, specifically the aspect ratio between the diameter and length of rod-like nanocrystals, played an important role in this phenomenon. (J. Schrier and K. B. Whaley, paper 1). A second study was carried out on InAs quantum dots, in collaboration with the group of G. Klimeck at JPL, Pasadena.

2. Subsequently in the second year of the program, these microscopic g-factor calculations were extended to a time-dependent method, which included the ability to treat the influence of finite electric fields. This study was able to quantitatively reproduce the experimental measurements, and to confirm the role of aspect ratio on the g-factor of this system. (P. Chen and K. B. Whaley, paper 2). In addition, the ability to manipulate the spin properties using applied electrical fields was used to provide insight into ongoing work on quantum control in the Whaley group. We are currently working to extend this method to the time-dependent treatment of magnetoexcitons (J. Schrier, P. Chen, and K. B. Whaley).

3. Additionally in the second year, we examined the magnetic properties of the Mn:ZnS dilute magnetic semiconductor nanocrystals. While the luminescence properties of the Mn centers have been studied extensively, the role of nanocrystal size and surface on the magnetic interactions between Mn centers is a topic of current interest. We found that experimental magnetic susceptibility results, which were used to suggest changes in the antiferromagnetic Heisenberg coupling constant between Mn atoms, could in fact be quantitatively reproduced by assuming inhomogeneous metal doping in the nanocrystals, a phenomenon that has been observed experimentally for these systems using TEM and chemical studies. (J. Schrier and K. B. Whaley, paper 3).

4. In the third year, we developed an atomistic treatment of the coherent spin transfer between nanocrystals linked by organic molecules. This was motivated by experimental TRFR studies indicating coherent spin transfer of approximately 20% at room temperature. Our results agreed qualitatively with the experimental measurements for the 1,4-dithiolbenzene and 1,4-dithiolcyclohexane molecules; additionally we used our method to study the role of molecular conformations and nanocrystal surface site attachments on the spin transfer, and to suggest new linking molecules that would utilize quantum interference effects to test the coherent through-molecule transport hypothesis. (J. Schrier and K. B. Whaley, paper 7).

5. In the third year, we also performed density-functional calculations on the endohedral metallofullerene, Gd@C82, in order to determine the structural, magnetic, and electronic transport properties for applications to future devices. The ground state geometry we calculated agrees with recent synchrotron powder-diffraction structure information, placing the Gd atom at a unique site compared to the other rare-earth metallofullerenes. Placement of the Gd atom at this position results in agreement with the magnetic properties determined by ESR and magnetic susceptibility experiments, in contrast to previous computational studies. Modeling a possible molecular electronics device by placing the Gd@C82 between two gold contacts, we find no spin-filter behavior as a result of interaction with the Gd spin. Instead, we find that the electron transport is dominated by the conjugated pi-electron system of the fullerene, which is disrupted by endohedral inclusion, reducing the conductivity. We are currently performing calculations on the magnetic and transport properties of (Gd@C82)@SWNT systems, in order to provide insight into recent experiments on these systems. (L. Senapati, J. Schrier, and K. B. Whaley, paper 6).

6. We have made microscopic calculations of the hyperfine coupling constants of endohedrally enclosed nitrogen atoms and the scaling of these with fullerene size, as required for implementation of quantum cellular automata proposals for quantum information processing using endohedral fullerenes. (J. Schrier and K. B. Whaley, paper 8).

List of Publications
Listings of all publications under this grant
(author, title, journal, issue, date)

* Papers published in peer-reviewed journals:
1. J. Schrier and K. B. Whaley, "Tight-binding g-factor calculations of CdSe nanostructures", Phys. Rev. B. **67**, 235301 (2003).

2. P. Chen and K. B. Whaley, "Magneto-optical response of CdSe nanostructures", Phys. Rev. B. **70**, 045311 (2004).

3. J. Schrier and K. B. Whaley, "A Simple Model for Magnetization Ratios in Doped Nanocrystals", J. Appl. Phys. **93**, 1436 (2004).

4. L. Senapati, J. Schrier, and K. B. Whaley, "Electronic Transport, Structure, and Energetics of Endohedral Gd@C82 Metallofullerenes", Nano Lett. **4**, 2073 (2004).

* Papers published in non-peer reviewed journals: N/A

* Papers presented at meetings, not published in conference proceedings: N/A

* Manuscripts submitted, but not published:
5. L. Senapati, J. Schrier, and K. B. Whaley, "Spin Polarized Electron Transport in a Magnetically Coupled Molecular Wire", in preparation (2004).

6. J. Schrier and K. B. Whaley, "Atomistic Theory of Coherent Spin Transfer between Molecularly Bridged Quantum Dots", in preparation (2004).

7. J. Schrier and K. B. Whaley, "Thomas-Fermi Theory of the Hyperfine Constants of Endohedral Fullerene Atoms", submitted J. Chem. Phys. (2004).

List of Participating Scientific Personnel and Degrees Earned
K. Birgitta Whaley, Professor
Joshua Schrier, graduate student (Ph.D. expected, Spring 2005)
Pochung Chen, postdoctoral fellow
Seungwon Lee, postdoctoral fellow
Lax Senapati, postdoctoral fellow

Report of Inventions
N/A